Appendix 2

Extended methods

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1. Laser ablation inductively coupled plasma mass spectrometer analysis of olivines

A survey of 162 olivines was conducted by laser ablation inductively coupled plasma mass spectrometry using a VG PQ Excell mass spectrometer interfaced with a NewWave 193 nm ArF Eximer laser at Lamont-Doherty Earth Observatory. Olivine (0.5-1mm diameter) was handpicked, mounted in dental resin, ground to expose a center-cut, and polished to 1 μm. Line scans were performed from core to rim at 3 μm/s using a beam with a 25 μm diameter and 15 Hz pulse frequency at 100% power (>10 J/cm^2 and >2 GW/cm^2). Traverses were preceded by a pre-ablation scan using a 40 μm beam at 10% power. BHVO-1, BIR-1, and GOR-132 were used as calibration standards. MgO was used as the internal standard, and elemental concentrations were obtained by assuming olivine stoichiometry.

2. Scanning electron microscope analysis olivines

For the 85 select olivines and the 53 olivine hosts of melt inclusions, we obtained backscattered electron (BSE) images using a Zeiss EVO60 variable pressure scanning electron microscope (SEM) at the American Museum of Natural History (AMNH). Dental resin mounts, polished to 0.25 μm, were carbon coated for BSE imaging and uncoated for electron backscatter detection (EBSD) analyses. BSE images were conducted using a 15 KeV, 30 μA focused beam at a working distance of 10-11 mm. For zoned crystals, we obtained 2 grayscale intensity profiles measured perpendicular to crystal edges (indicated by the presence of adhering matrix glass) using ImageJ software. EBSD analyses were conducted using a 20 KeV, 40 μA focused beam at a

working distance of 10-15 mm. Angles between selected profiles and principal axes *A*, *B*, and *C* were calculated using Stereonet 9.5 software (Cardozo and Allmendinger, 2013).

3. Electron microprobe analysis of olivines

Quantitative measurements of olivine compositions were completed using a Cameca SX-100 electron microprobe (EMP) at AMNH. For zoned olivine, line scans of 4 to >10 points were collected to calibrate BSE images (Martin et al., 2008). Olivine hosts were also analyzed with ~3 replicate point analyses, conducted ~50 μm from the inclusions. San Carlos olivine was used as a check standard (Appendix C). Mg, Si, Mn, Ca, and Fe were analyzed using a 1 μm beam with a 15 KeV accelerating voltage and 10 nA current with on-peak times of 20 s and off-peak times of 10 s.

4. Raman spectroscopic analysis of vapor bubbles

We examined 53 olivine-hosted melt inclusions, 25 containing vapor bubbles. The density of CO_2 in the bubbles was calculated from Raman spectra obtrained using a JY Horiba LabRam HR (800 mm) spectrometer connected to a 514 nm Laser Physics 100s-514 Ar laser at Virginia Tech, following Lamadrid et al. (2017). Each bubble was analyzed 3 times in the center to assess precision and once near the edge check for the presence of carbonate that couldn't be visually detected. Of the 18 analyzed vapor bubbles, none appeared to contain carbonate and 11 exhibited a measurable fermi diad. Fermi diad peak positions were determined using a mixed Gaussian-Lorenzian peak-fitting routine in Thermo Galactic Grams. Inclusion and vapor bubble volumes were determined assuming ellipsoidal and spherical geometry, respectively. For inclusions, the axis perpendicular to the section was assumed to be equal in length to the short axis in cross section (Moore et al., 2015)

5. Fourier transform infrared spectroscopy of glasses

We measured H_2O and CO_2 contents in 49 melt inclusions and 12 matrix glasses using a Thermo-Nicolet Nexus 670 FTIR spectrometer coupled with a Continuum IR microscope at AMNH. In most cases, 3 replicate analyses were performed. Concentrations were calculated using the Beer-Lambert law, which depends on absorption coefficient, measured absorbance, and wafer thickness. Total H_2O concentration was determined using the peak centered around 3520 cm^{-1} with an absorption coefficient of 63 L/mol•cm (Dixon et al., 1997). CO_2 concentration was calculated by averaging the absorbance values of the carbonate peaks at 1430 and 1515 cm^{-1} and using the compositionally-dependent absorptivity formulation of Dixon and Pan (1995). For both, the background was determined using a cubic spline interpolation. Wafer thickness was measured using the reflectance technique of Wysoczanski and Tani (2006), and results were tested against tabletop micrometer measurements, which agreed within error.

6. Electron microprobe analysis of glasses

Major elements and volatiles (S, Cl) were measured in melt inclusions and matrix glasses on the CAMECA SX-100 at AMNH. We performed 3 replicate analyses near the center of each inclusion, taking care to avoid any interaction with the olivine crystal and previously analyzed regions. Glass 892-1 was used as a check standard (Appendix C). The beam was set to a 10 μm diameter, 15 kV accelerating voltage, and currents of 10 nA (for Na, Si, Mg, Al, K, Ca, and Fe) and 50 nA (for P, S, Cl, Ti, Mn). Count times were 5 s (Na), 20 s (Si, Mg, Al, K, Ca, S, Ti), 30 s (Cl, Mn), 40 s (P), and 50 s (Fe).

7. Shear-wave splitting data and analytical approach

For shear-wave splitting (SWS) analysis, we employ a semiautomated method (Savage et al., 2010a) based on the Shear wave Birefringence Analysis (SHEBA)/Silver and Chan algorithms (Teanby et al., 2004; Silver and Chan, 1991), which analyzes multiple time windows around a

manual S-wave time pick (we re-picked all S-wave arrival times for this study) and performs cluster analysis to determine and automatically grade the best estimate of Φ (fast wavelet polarization, a proxy for the orientation of maximum horizontal compression) and delay time (dt; a proxy for path length through the anisotropic volume and/or magnitude of the anisotropy). The Savage et al. (2010a) method first applies a set of user-defined filters to the earthquake waveform to find the three frequency bands with the highest signal-to-noise ratios. Each waveform is then analyzed up to three times (once for each of the three best filters that produce a reasonable signal-to-noise ratio), and may contribute only the best of three splitting measurements.

For our analysis, we focus on the AVO short-period three-component station SSLS, which is ~5 km from the vent. Continuous seismic waveform data from SSLS were available beginning in January 1999, and a catalog of detected (primarily volcanic) event waveforms was available for 1998. We cross-referenced these waveform data with the Alaska Earthquake Center (AEC; formerly AEIC) catalog of regional seismic events to obtain as large a set as possible of event waveforms for regional earthquakes in 1998-1999 that had epicenters within 100 km of Shishaldin and depths of less than 100 km. These selection criteria, combined with available waveform data, resulted in 46 analyzed earthquakes for the period July 1998-December 1999.

We rely on a strict two-level grading scheme to evaluate the results of SWS analysis: Automated assignment of an A, B, C, or D grade is based on the clustering properties of the solutions found using a set of slightly different analysis windows around the user-specified S wave pick (Savage et al., 2010a, 2010b). Specifically, the code compares all identified clusters of solutions and assigns a grade based on the uniqueness of the best cluster solution (for details, see Savage et al. (2010a)). The automated grading scheme also marks measurements with ambiguous "NULL" polarizations (these were discarded). Measurements which received an A or B grade in

automated grading were then manually evaluated and assigned a second A–D grade based on stability and average error of the solutions over different time windows, evidence of cycle skipping, similarity between the two rotated and transformed S-wavelets, and linearity of the resolved (unsplit) S-wave particle motion (e.g., Gerst and Savage, 2004). In this study, we present and analyze only the 11 SWS measurements, of individual earthquakes, assigned an A or B grade in both automated and manual grading.

References

Cardozo, N., Allmendinger, R.W., 2013. Spherical projections with OSXStereonet. Computers & Geosciences 51, 193-205.

Dixon, J.E., Clague, D.A., Wallace, P., Poreda, R., 1997. Volatiles in Alkalic Basalts form the North Arch Volcanic Field, Hawaii: Extensive Degassing of Deep Submarine-erupted Alkalic Series Lavas. Journal of Petrology 38, 911-939.

Gerst, A., Savage, M.K., 2004. Seismic anisotropy beneath Ruapehu Volcano: a possible eruption forecasting tool. Science 306, 1543–1547.

Lamadrid, H.M., Moore, L.R., Moncada, D., Rimstidt, J.D., Burruss, R.C., Bodnar, R.J., 2017. Reassessment of the Raman CO_2 densimeter. Chemical Geology 450, 210-222.

Dixon, J.E., and Pan, V. (1995) Determination of the molar absorptivity of dissolved carbonate in basanitic glass. American Mineralogist, 80, p. 1339.

Martin, V.M., Morgan, D.J., Jerram, D.A., Caddick, M.J., Prior, D.J., Davidson, J.P., 2008. Bang! Month-Scale Eruption Triggering at Santorini Volcano. Science 321, 1178-1178.

Moore, L.R., Gazel, E., Tuohy, R., Lloyd, A.S., Esposito, R., Steele-MacInnis, M., Hauri, E.H., Wallace, P.J., Plank, T., Bodnar, R.J., 2015. Bubbles matter: An assessment of the

contribution of vapor bubbles to melt inclusion volatile budgets. American Mineralogist 100, 806-823.

Teanby, N., Kendall, J.-M., van der Baan, M., 2004. Automation of shear-wave splitting measurements using cluster analysis. Bull. Seismol. Soc. Am. 94, 453–463.

Savage, M.K., Wessel, A., Teanby, N.A., Hurst, A.W., 2010a. Automatic measurement of shear wave splitting and applications to time varying anisotropy at Mount Ruapehu volcano, New Zealand. J. Geophys. Res. 115, B12321, http://dx.doi.org/10.1029/2010JB007722.

Savage, M.K., Ohminato, T., Aoki, Y., Tsuji, H., Greve, S., 2010b. Absolute stress and its temporal variation at Mt. Asama Volcano, Japan, from seismic anisotropy and GPS. Earth Planet. Sci. Lett. 290, 403–414.

Silver, P.G., Chan, W.W., 1991. Shear wave splitting and subcontinental mantle deformation. J. Geophys. Res. 96, 16429–16454.

Wysoczanski, R., Tani, K., 2006. Spectroscopic FTIR imaging of water species in silicic volcanic glasses and melt inclusions: An example from the Izu-Bonin arc. Journal of Volcanology and Geothermal Research 156, 302-314.